



Evaluation of Inhalation Exposure to Carcinogenic PM₁₀-Bound PAHs of People at Night Markets of an Urban Area in a Metropolis in Eastern China

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ABSTRACT

Inhalable particle bounded carcinogenic polycyclic aromatic hydrocarbons (PAHs) pose high health risks to both food service workers and consumers cooking or purchasing late night snacks, respectively, in night markets because PAHs are generated in abundance during thermal cooking processes such as barbecue grilling. In the present study, sixteen carcinogenic PAHs in PM₁₀ collected during open hours from eight night markets in an urban area of a metropolis in eastern China were determined. The total concentration of PAHs (Σ PAH) ranged from 145 to 1340 ng m⁻³, with an arithmetic mean of 828 ± 360 ng m⁻³. Five- and six-ringed PAHs were predominant in the composition, implying a combination of sources including cooking and traffic. Low coefficient of divergence values for individual PAH homologous among all sampling sites indicated similar sources. The occurrence of PM₁₀-bound PAHs appeared to depend on the gas-particle partitioning processes and emission sources according to the results of principal component analysis (PCA). The daily inhalation rate of particle and predicted gaseous benzo[a]pyrene (BaP)-equivalent adjusted PAHs (BaP_{eq}) for the workers due to occupational exposure was 0.451–3.43 μg day⁻¹. Correspondingly, the occupational exposure risk for workers with one year of service was less than the acceptable risk level (10⁻⁶). However, the risk for people who had worked for more than 3 years exceeded 10⁻⁶ but was less than the priority risk level (10⁻⁴). For consumers, the maximum consumption time (t_{\max}) for each time under the acceptable risk level would increase with an increase in age, and the exposure risk for infants, toddlers and children is high when PM₁₀-bound PAHs are inhaled. We recommend that t_{\max} should be less than 1 hour for consumers under 25 years old. Therefore, our results indicate that both workers and consumers in the night markets have high cancer risks due to inhalation of PM₁₀-bound PAHs.

Keywords: Polycyclic aromatic hydrocarbons (PAHs); PM₁₀; Exposure risk; Night markets.

INTRODUCTION

Cooking and food processing are known to generate inhalable particle and various toxicants (Jägerstad and Skog, 2005). Among many mutagenic and carcinogenic compounds, polycyclic aromatic hydrocarbons (PAHs) are gaining wide public concern because these multi-ringed organic compounds are one of the first verified airborne carcinogens; some PAH congeners have been classified as "carcinogenic to humans" (Group 1) or "probably carcinogenic to humans" (Group 2A and 2B) (International Agency for Research on Cancer (IARC), 2012). For example, benzo[a]pyrene (BaP) and dibenz[a,h]anthracene (DahA) are listed in Group 1 and Group 2A, respectively. Benz[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), and benzo[k]fluoranthene

(BkF) are the members of Group 2B. Moreover, PAHs are heavily produced from various types of cooking sources via the incomplete combustion of organic materials (See *et al.*, 2006; Farhadian *et al.*, 2010), and cooking processes usually generate PAHs with high carcinogenic potency. Li *et al.* (2003) estimated that the total emission inventory for 21 PAHs from cooking sources (8,973 kg year⁻¹) was significantly less than that from traffic sources (13,500 kg year⁻¹) in a city of southern Taiwan. However, the toxic equivalency factor-adjusted emission rates (BaP was treated as a reference compound) from cooking sources (675 kg year⁻¹) were more than those from traffic origins (61.4 kg year⁻¹) (Li *et al.*, 2003). Therefore, PAHs from incomplete combustion of organic materials during thermal cooking pose inevitable stressors to both occupational and public health (Kuo *et al.*, 2005; Shen *et al.*, 2011).

Chinese cooking methods and barbecue grilling with liquefied petroleum gas and charcoal as fuels generate large quantities of PAHs (Li *et al.*, 2003; Chen *et al.*, 2012). On one hand, these toxicants are contained the cooked foods, and consumers increase their cancer risk by dietary intake

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(Alomirah *et al.*, 2011; Kao *et al.*, 2014). On the other hand, PAHs can be released into air within gaseous and particulate phases, and workers and consumers increase their cancer risk by inhalation (Kuo *et al.*, 2006; Chen *et al.*, 2012). Carcinogenic PAHs with high molecular weights apparently tend to bind to particles due to their low volatilities (Shen *et al.*, 2014). PAHs associated with particles having aerodynamic diameters below 10 μm (regarded as PM_{10}) can deposit into the human respiratory system because PM_{10} is inhalable.

Nevertheless, most previous works have demonstrated indoor air pollution caused by PAHs from cooking processes (Bhargava *et al.*, 2004; Ding *et al.*, 2012; Shen *et al.*, 2014). Limited studies regarding PAHs from outdoor cooking, e.g., barbecue stalls providing late night snacks in the night markets, have been published (Zhao *et al.*, 2011). Currently, late night snacks, primarily grilled foods, are extremely popular in urban areas around China, and many well-known snack streets or squares in almost every city are composed of several barbecue stalls. Therefore, PM_{10} -bound PAHs from barbecue grilling at night markets should be of great concern to protect the workers' and consumers' health.

The aims of the present study were as follows: (1) to investigate the occurrence of PM_{10} -associated carcinogenic PAHs at the night markets in a metropolis in eastern China, (2) to estimate the daily inhalation dosage of target PAHs and to evaluate the potential cancer risk by inhalation for

food service workers in the night markets, and (3) to provide the maximum consumption time for consumers.

MATERIALS AND METHODS

Sampling Sites

Eight sampling sites (Fig. 1) total were selected in the urban area of Hefei, a metropolis in eastern China with a total population of approximately 2.2 million. Except for the site located in the northern gate of the western campus of USTC where many barbecue stalls were closed because of summer vacation for university students, seven sites were night market areas with dense food stalls where barbecue grilling was the primary cooking method (Table 1). Liquefied petroleum gas was commonly used for fuel. Approximately three workers were in each stall, and dining tables for consumers were behind the stalls. Furthermore, the night markets were near the traffic roads, and no factories were near the eight sampling sites. Additionally, a control site in a rural area in southwestern Hefei, which was far from any night market, was selected to collect PM_{10} samples for comparison with the results from the night markets.

Air Sampling and Metrological Conditions

According to the questionnaire before our sampling, cooking in the market is performed primarily from 20:00 h to 2:00 h. Thus, the sampling was conducted at night from

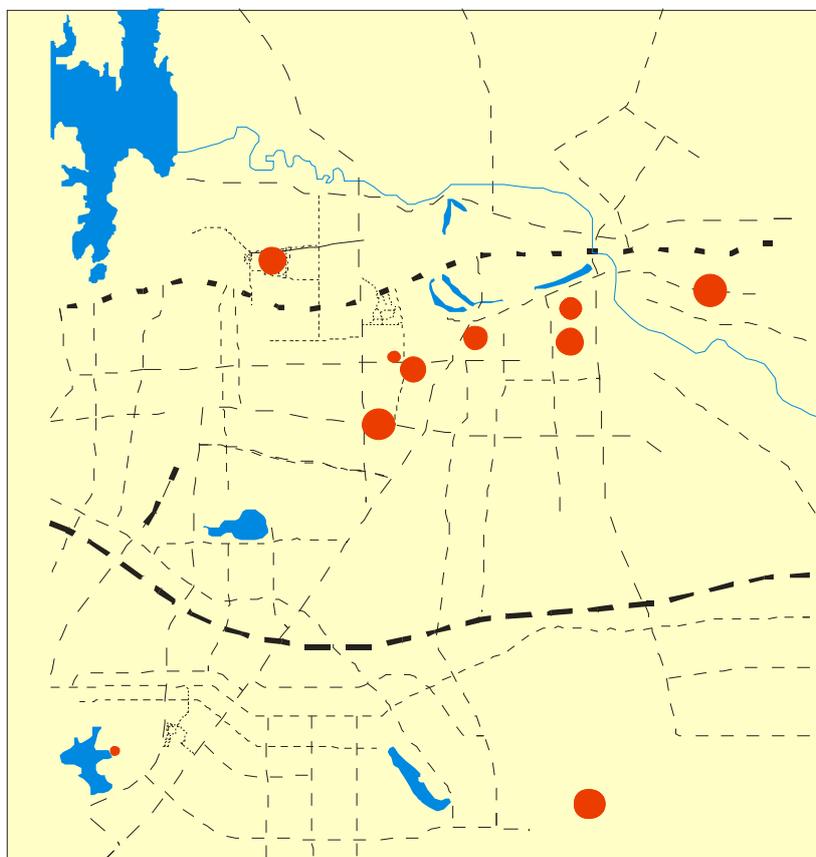


Fig. 1. Sampling sites and spatial distribution of PM_{10} -bound ΣPAH concentrations. The sizes of the symbols are proportional to the concentration values.

Table 1. Sampling information.

	Northern gate of western campus of USTC	Eastern gate of western campus of USTC	Xuancheng Road	Tunxi Road	Susong Road	Pihe Road	Datong Road	Guogou Square
latitude	31.843°N	31.841°N	31.854°N	31.848°N	31.845°N	31.852°N	31.860°N	31.857°N
longitude	117.251°E	117.255°E	117.290°E	117.291°E	117.273°E	117.255°E	117.313°E	117.235°E
sampling date	2012/7/16	2012/7/17	2012/7/18	2012/7/19	2012/7/20	2012/7/21	2012/7/22	2012/7/23
wind speed	3.4–7.9	< 5.4	< 5.4	3.4–7.9	3.4–7.9	< 5.4	< 5.4	3.4–7.9
prevailing wind direction	E	NE	No constant wind direction	E	S	S	No constant wind direction	SE
average air temperature	24	25	24	24	26	27	28	28
concentration of PM ₁₀ (μg m ⁻³)	246.3	356.6	172.8	227.9	169.1	426.5	356.6	176.5
surrounding information	No barbecue stalls, surrounded by main traffic road (Huangshan Road)	5–7 barbecue stalls	5–7 barbecue stalls	4–5 barbecue stalls, surrounded by main traffic road (Tunxi Road)	4–5 barbecue stalls	5–7 barbecue stalls	> 20 barbecue stalls	5–7 barbecue stalls

20:00 h to 2:00 h. PM₁₀ samples were collected by air samplers (ZC-Q0102, Zhejiang Hengda Instrument & Meter Co., Ltd., Zhejiang, China) with a Whatman glass fiber filter (GFF, 102 mm i.d.). All of filters were pre-heated at 450°C for 5 hours, then wrapped in pre-baked aluminum foil packages, sealed in airtight polyethylene bags and reserved in a desiccator. Before the sampling, the filters were weighed. The samplers were set at a height of approximately 1.5 m above the ground within the breathing zone. The airflow was set to approximately 90 L min⁻¹. After the sampling, the GFFs were put into refrigerator at -4°C immediately and were ready for the following treatment.

Information regarding the sampling is presented in Table 1. Briefly, the air temperature was approximately 24–29°C during the summer sampling period from July 16 to 23, 2012, and the wind strength was under 3–4 Beaufort wind force scale.

Materials and Reagents

Pesticide grade hexane, methylene chloride and methanol were obtained from Oceanpak Alexative Chemical Company (Gothenburg, Sweden). Neutral silica gel (80–100 mesh) obtained from Guangzhou Xinshi Chemistry Experimental Equipment (Guangzhou, China) was successively pre-cleaned by Soxhlet extraction for 48 h by methanol and methylene chloride before use. Sodium sulfate obtained from Sigma-Aldrich (St. Louis, MO, USA) was baked at 450°C for 4 hours and stored in sealed containers. All glassware was cleaned with methanol, chromic acid and tap water, followed by rinsing with deionized water, and then baked at 150°C overnight. The cleaned glassware was covered with aluminum foil and stored at room temperature before use. A stock mixture solution containing 16 carcinogenic PAH compounds, namely, phenanthrene (Phe), anthracene (Ant), 2-methylphenanthrene (2-MPhe), 1-methylphenanthrene (1-MPhe), fluoranthene (Flu), pyrene (Pyr), BaA, Chr, BbF, BkF, benzo[e]pyrene (BeP), BaP, perylene (Per), indeno[1,2,3-cd]pyrene (IcdP), DahA, and benzo[g,h,i]perylene (BghiP) was purchased from AccuStandard (New Haven, CT, USA). Three deuterated standards including five PAHs (phenanthrene-*d*₁₀, chrysene-*d*₁₂, and perylene-*d*₁₂ at 100 μg mL⁻¹ each; Cambridge Isotope Laboratory Inc., Massachusetts, USA) were used as surrogate standards to monitor the performance of sample preparation processes for PAHs. 2-Fluoro-1,1-biphenyl and *p*-terbenyl-*d*₁₄ were employed as internal standards to quantify PAHs.

Extraction and Clean up

Before Soxhlet extraction, each sample was spiked with the surrogate standards. The GFFs were extracted with a 1:1 mixture of hexane and dichloromethane for 48 hours. The extracts were concentrated to approximately 1 mL using a rotary evaporator (ZX98-1, Shanghai Yukang), and this step was repeated after the solvent was exchanged to 10 mL hexane. Then, the samples were cleaned using a chromatographic column packed with pre-extracted cotton, 12 cm silica (180°C baked) and 2 cm anhydrous sodium sulfate from the bottom to the top. The fraction containing

PAHs was eluted by 15 mL of hexane:dichloromethane mixture (7:3 in volume). The elute was concentrated to 1 mL using a rotary evaporator, and then the solvent was exchanged to hexane and was further reduced to 0.5 mL with a soft stream of purified nitrogen. The final samples were spiked with the internal standards and stored at -4°C until they were analyzed.

Instrumental Analysis

A gas chromatograph-mass spectrometer (GCMS-QP2010 Plus supplied by Shimadzu Scientific Instruments, Kyoto, Japan) was employed to quantify the individual concentrations of 16 PAH congeners. A DB-5MS (length = 60 m; inner diameter = 0.25 mm; film thickness = 0.25 μm) column was used for chromatographic separation. The initial oven temperature was set at 60°C and programmed at a rate of $10^{\circ}\text{C minute}^{-1}$ to 200°C , at a rate of $2^{\circ}\text{C minute}^{-1}$ to 250°C , and finally at a rate of $20^{\circ}\text{C minute}^{-1}$ to 290°C and then kept at 290°C for 5 minutes. The injection port temperature was programmed from 100°C to 280°C at a rate of $200^{\circ}\text{C minute}^{-1}$ and kept at 280°C for 10 minutes. Each sample (1 μL) was injected using an automatic AOC-20 Series sampling system in the splitless mode. One minute after injection, the split mode was switched on with a split ratio of 100:1, and five minutes later, the ratio was changed to 20:1 to save the carrier gas. The ion source and interface temperatures were both maintained at 250°C .

Quality Assurance

Three field blanks were applied to monitor the background contamination and were processed sequentially with field samples. Three method blanks were also analyzed. These contaminants did not interfere with our targets. Surrogate standards were added into each sample as the recovery indicators. The average recovery percentages of these substances were $113 \pm 12\%$, $117 \pm 10\%$, and $99 \pm 13\%$ (mean \pm relative standard deviation, $n = 14$).

Data Analysis

All measured concentrations of particulate PAHs were not adjusted by the recovery efficiencies. The total concentration of the 16 PAHs was defined as ΣPAH . The analysis of the correlation between the concentrations of ΣPAH and PM_{10} was performed using two-sided Spearman's correlation at a significance level of 0.05, and principal component analysis of the mass concentrations of individual PAH compounds was performed using SPSS 16.0 software package (SPSS Inc., Chicago, IL, USA).

(1) Coefficient of divergence (CD)

To evaluate the spread of the data points for two datasets, the coefficient of divergence (CD) was employed as described in Eq. (1):

$$CD_{jk} = \sqrt{\frac{1}{p} \sum_{i=1}^p \left(\frac{C_{ij} - C_{ik}}{C_{ij} + C_{ik}} \right)^2} \quad (1)$$

where i and j are the two profiles of the sampling sites, p is the

number of components; and C_{ij} and C_{ik} are the concentrations of component i at sampling sites j and k , respectively.

(2) Estimation of gaseous concentrations of individual PAH congeners

To estimate the gaseous concentrations of individual PAH congeners, the particle-gas partition coefficient (K_p , with a unit of $\text{m}^3 \mu\text{g}^{-1}$) was applied as described elsewhere (Cincinelli et al., 2014):

$$K_p(i) = \frac{C_p(i)}{C_g(i) \times \text{PM}_{10}} \quad (2)$$

where $C_p(i)$, and $C_g(i)$ are the concentrations of PAH congener i in PM_{10} and in gaseous phase (ng m^{-3}), respectively, and PM_{10} is the concentration of PM_{10} (ng m^{-3}). K_p of each compound generally is a function of its subcooled liquid vapor pressure (P_L) at 25°C , and $\log K_p$ can be linearly correlated to the $\log P_L$ as shown in Eq. (3) as proposed by Simcik et al. (1998). P_L values ranged from 9.51×10^{-8} Pa for DahA to 0.115 Pa for Phe (Paasivirta et al., 1999)

$$\log K_p = -1.1 \log P_L - 5.28 \quad (3)$$

(3) Inhalation exposure and health risk assessment

The inhalation exposure of the PAH mixture was adjusted by the toxicity equivalent factor (TEF) to BaP toxicity equivalent (BaP_{eq}) of each compound (Nisbet and LaGoy, 1992).

$$\text{BaP}_{\text{eq}} = \sum (C_p(i) + C_g(i)) \times \text{TEF}_i \quad (4)$$

The daily inhalation exposure level (IEL) of PAHs for the occupational staff was calculated by the BaP_{eq} concentration, inhalation rate (IR, $\text{m}^3 \text{h}^{-1}$) and daily working time (t) in the following equation:

$$\text{IEL} = \text{BaP}_{\text{eq}} \times \text{IR} \times t \quad (5)$$

Considering that the vendors in the night market are adults (usually ages ranging from 20–50 years old), the IR was selected at $1.6 \text{ m}^3 \text{h}^{-1}$ (Duan, 2012), and the daily work time was set at 6 hours (from 20:00 h to 2:00 h).

The incremental lifetime cancer risk (ILCR) of vendors by inhalation exposure to ambient air PAHs was computed by Eq. (6):

$$\text{ILCR} = \frac{\text{SF} \times \text{IEL} \times \text{EF} \times \text{T} \times \text{CF}}{\text{BW} \times \text{AT}} \quad (6)$$

where SF is the cancer slope factor for BaP inhalation exposure (a geometric mean of $3.14 \text{ kg day mg}^{-1} \pm$ a geometric standard deviation of 1.80) (Chen and Liao, 2006), EF is the exposure frequency (day year^{-1} , with 250 d year^{-1} selected in the present study), T is the exposure duration (year, with 1, 3, 5, 10 and 20 years selected to evaluate the risk of the vendors with 1, 3, 5, 10, and 20 years of seniority, respectively), CF is the conversion factor (10^{-6}

mg ng⁻¹), BW is the body weight of the vendors (67.1 ± 11.4 kg) (Xia *et al.*, 2013), and AT represents the average lifespan for carcinogens (25550 days) (Xia *et al.*, 2013).

(4) Estimation of maximum consumption time (t_{\max})

To estimate t_{\max} in the barbecue night market for consumers under acceptable ILCR (ILCR_{acceptable}) at 10⁻⁶, we hypothesized consumption frequency in the barbecue night market at once per week before 55 years old with the lifespan for carcinogens of 70 years old (25550 days). The maximum consumption time for each time in the night market could be calculated by Eq. (7):

$$t_{\max} = \frac{\text{ILCR}_{\text{acceptable}} \times \text{BW} \times \text{AT}}{\text{SF} \times \text{BaP}_{\text{eq}} \times \text{IR} \times \text{EF} \times \text{T} \times \text{CF}} \quad (7)$$

RESULTS AND DISCUSSION

PM₁₀ and PAH Concentrations

The level of PM₁₀ in the present study ranged from 168 to 426 μg m⁻³, with an arithmetic mean ± deviation of 267 ± 100 μg m⁻³, while the concentration of PM₁₀ at the control site was 55.1 μg m⁻³. The mean concentration of PM₁₀ was 2-fold higher than that observed at a night market for barbecue (Kuo *et al.*, 2005). Compared with other occupational circumstances, field traffic police personnel (Majumder *et al.*, 2012) and poultry farming (Lawniczek-Walczuk *et al.*, 2013) have a greater risk of exposure to PM₁₀; however, the employees of public transportation (Gerber *et al.*, 2014), lignite-fired power plant (Manousakas *et al.*, 2013), and Sports facilities (Alves *et al.*, 2013) had lower exposure to PM₁₀. All values obtained in the present study exceeded the primary national ambient air quality standard of China for PM₁₀ (150 μg m⁻³) (Ministry of Environmental Protection of the People's Republic of China, 2012). Therefore, PM₁₀-related airborne particulate matter exposure is considered an important environmental stressor for vendors of the night markets because numerous studies have confirmed the negative effects of chronic and acute exposure to PM₁₀ on human health (Pope *et al.*, 1991; Giri *et al.*, 2007).

The mean concentration of ΣPAH was 828 ± 360 ng m⁻³, with a range from 145 to 1340 ng m⁻³ at night markets and 101 ng m⁻³ at control site. The maximum concentration of ΣPAH was observed at Datong Square, which has more than 20 barbecue stalls and which is the most popular barbecue night market in Hefei City. The lowest concentration of ΣPAH was investigated at the northern gate of the western campus of the University of Science and Technology of China. During the sampling period, most barbecue stalls were closed because of summer vacation for university students. Furthermore, the concentrations of PAHs in the present study were significantly lower than those investigated in the barbecue night markets that used charcoal as fuel, where the concentration of 22 PAH congeners ranged from 1690 to 31000 ng m⁻³. However, our results were comparable with the values of PM₁₀-bound PAH concentrations found in the barbecue night markets that used electricity as fuel (Kuo *et al.*, 2005) and with the values of PM_{2.5}-bound PAH

concentrations found in the barbecue night markets that used liquefied petroleum gas as fuel (Zhao *et al.*, 2011). The concentrations of ΣPAH in the present study were also lower than those observed in suburban areas (Jamhari *et al.*, 2014) and in high-density traffic areas (Chen *et al.*, 2013). The low concentrations of PM₁₀-bound PAHs found in the barbecue night markets in Hefei City were most likely due to the prohibition of charcoal as fuel for barbecue (Wu *et al.*, 2012). PAHs could be formed both during cooking and food processes (Onyango *et al.*, 2012) and fuel combustion (Hou *et al.*, 2008). Previous studies have demonstrated that the concentrations of PAHs in grilled foods depend on the thermal processes and grilling methods (Farhadian *et al.*, 2010; Viegas *et al.*, 2012). Generally, the concentrations of PAHs in charcoal-grilled foods are significantly higher than those in gas-grilled foods and electric oven-grilled foods (Farhadian *et al.*, 2010). Therefore, fewer PAH emissions are expected when grilling foods with liquefied petroleum gas as fuel compared with charcoal as fuel. Furthermore, liquefied petroleum gas combustion during cooking causes lower concentrations of ambient PAHs than does charcoal consumption (Titcombe and Simcik, 2011). However, the PM₁₀-bound PAH concentrations in the barbecue night markets in Hefei City were 1–2 orders of magnitude higher than those detected in urban areas (Mantis *et al.*, 2005; Hong *et al.*, 2007; Vu *et al.*, 2011; Limu *et al.*, 2013; Wu *et al.*, 2014a), indicating that staff and consumers in barbecue night markets are significantly exposed to carcinogenic PAHs.

Two-tailed Spearman's correlation analysis indicated that the concentrations of ΣPAH did not correlate to the content of PM₁₀ ($R^2 = 0.31$ and $p = 0.45$), which is partly attributable to the effects of particle sizes because the distribution of airborne PAHs usually depends on the particle sizes (Kawanaka *et al.*, 2009; Zhang *et al.*, 2012) and because these semi-volatile organic pollutants with low vapor pressures are generally associated with ultrafine particles (Offenberg and Baker, 1999). A previous study demonstrated that most PAHs from grilled foods (corn, trout, beef, prawns, and pork) were observed in particles less than 0.43 μm in size (Saito *et al.*, 2014); however, PAHs were predominantly emitted within the 1.1–2.1 μm particles when grilling fishes (Tanaka *et al.*, 2012). A study regarding the PAH emissions from crop residues burned for cooking also indicated that particles with an aerodynamic diameter between 1.1–2.1 μm were capable of concentrating carcinogenic aromatic hydrocarbons (Shen *et al.*, 2011). Additionally, temperature is expected to affect the redistribution of PAHs between airborne particle matter and gases (Tsapakis and Stephanou, 2005), and wind speed also affects the concentration of particle-bound PAHs by the resuspension of street dust (Martuzevicius *et al.*, 2011). Although all samples were collected during the same time (between 20:00 h to 2:00 h), the air temperature, wind speed and wind direction slightly varied during the sampling period (Table 1) in the present study. Consequently, particle-bound PAHs were not distinctly dependent on the PM₁₀ content.

Compositional Characteristics

In the present study, the most abundant PAH was BbF

($15.0 \pm 2.47\%$), followed by BghiP ($14.4 \pm 2.49\%$), BeP ($10.8 \pm 1.92\%$), IcdP ($10.3 \pm 1.89\%$), and Flu (9.03 ± 3.16) (Fig. 2). The samples contained low concentrations of methylated Phe and Per, with the relative abundance of individual PAH compounds less than 4%. Low CD values (0.15–0.42) between each two sampling sites were investigated, reflecting all sampling sites with similar composition distributions and further implying similar sources of PM₁₀-associated PAHs for all sampling sites because a CD value close to zero generally suggests similarities in PAH sources among sites, while a CD value approaching 1 implies different PAH sources (Shi *et al.*, 2009; Kong *et al.*, 2010; Kong *et al.*, 2013).

A previous study indicated that BbF, IcdP, BeP were the most abundant PM_{2.5}-associated PAHs in Chinese cooking stalls (See *et al.*, 2006), whereas particle samples collected in the kitchens that used liquefied petroleum gas as fuel (See *et al.*, 2006) and in the night market (Zhao *et al.*, 2011) contained the highest observed concentrations of BghiP, IcdP, Flu, Pyr and BaA. Therefore, the PM₁₀-bound PAHs in the night markets of Hefei City were predominantly derived from cooking. Flu and Pyr are usually considered markers of traffic-related sources (Ho *et al.*, 2009). In the present study, high concentrations of Flu and Pyr also suggest vehicle-associated mobile sources because many gasoline- and liquefied petroleum gas-fueled vehicles were present around the night markets for consumer transport. Therefore, airborne PAHs in the night markets of Hefei City originated from combined cooking and vehicle emissions.

The gaseous concentrations of individual PAH congeners were estimated to range from 3970 ng m⁻³ for Phe to < 1 ng m⁻³ for heavy molecular weight PAHs (BbF, BkF, BaP, Per, IcdP, DahA and BghiP (Fig. 2). High concentrations of light molecular weight PAHs were predicted in the gaseous phase because of their volatilization. Previous studies have

indicated that light molecular weight PAHs predominantly occur in the gaseous phase, while heavy molecular weight PAHs tend to accumulate in the particle phase (Tasdemir and Esen, 2007; Vasilakos *et al.*, 2007). However, heavy molecular weight PAHs are likely to be underestimated in the gaseous phase because PAHs from cooking can occur in oil fumes, which are usually considered in gaseous phase. Therefore, high concentrations of heavy molecular weight PAHs were observed in gaseous phase at barbecue night market (Zhao *et al.*, 2011) and in kitchens (Chen *et al.*, 2012). It should be noted that the estimated gaseous concentrations of ΣPAH were 2.09–21.1 times higher than the measured PM₁₀-bound PAH concentrations, but BaP_{eq} concentrations in gaseous phase were significantly lower than those found in PM₁₀ samples. The primary reason is due to the fact that the predicted gaseous concentrations of ΣPAH were largely dominated by Phe and Ant (accounting for $84.4 \pm 5.3\%$), which are characterized by high volatilities and low toxicity equivalent factors when comparing with those high molecular weight PAHs.

Principal Component Analysis

In the present study, principal component analysis based on the particle-bound PAH data potentially classified PAH compounds into two groups (Fig. 3(a)), and the top two components explained 87.6% of the total variance, with component 1 (PC 1) = 60.0% and component 2 (PC 2) = 27.6%. PC 1 was associated primarily with the heavy weight molecular PAHs, including BbF, BkF, BeP, BaP, Per, IcdP, DahA, and BghiP, as well as Ant, with negative loading. This component was possibly affected by the volatilities of PAHs and gas-particle partitioning processes because the PC 1 loading of the rotated component matrix clearly correlated with the subcooled liquid vapor pressures (P_L^0) of PAH isomers (Fig. 3(b)). The partition processes of semi-organic

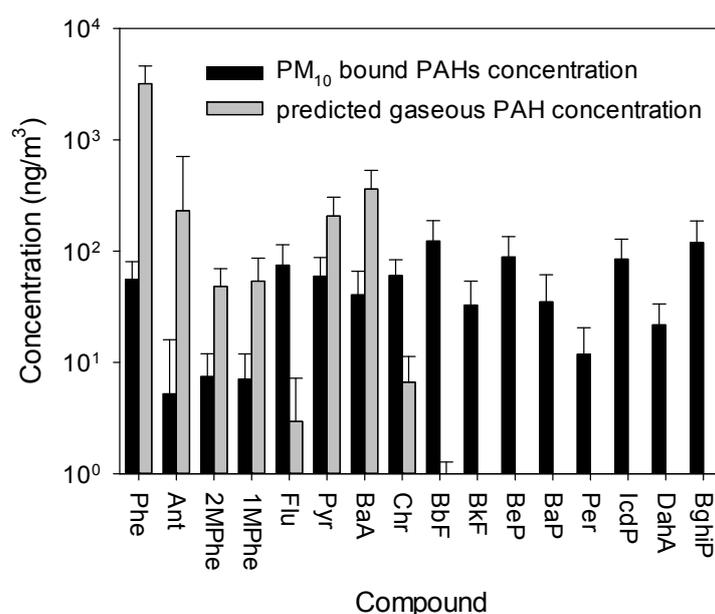


Fig. 2. Average concentrations for particulate (measured) and gaseous (predicted) phase PAHs in the night markets, Hefei City, eastern China.

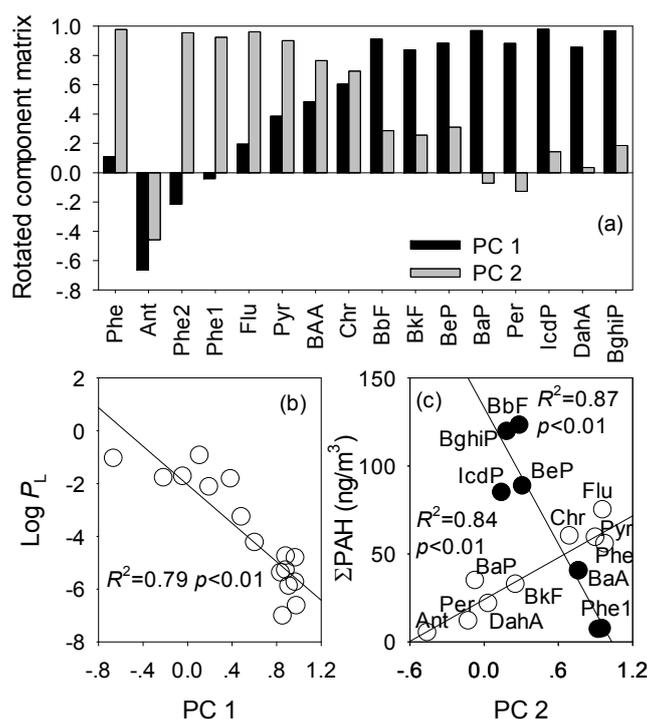


Fig. 3. Rotated component matrix of 16 PAH homologs in PM_{10} samples by principal component analysis (a); the loading of component 1 (PC 1) and 2 (PC 2) was plotted against the subcooled liquid vapor pressures (P_L°) of PAHs (b) and their mass concentrations (c).

contaminants between gas and particle phases are generally explained by physical adsorption onto the surface of particle matter or adsorption into the organic matter, both of which are associated with the P_L° (Simcik *et al.*, 1998). Consequently, numerous studies have demonstrated that the particle-gas partition coefficients of semi-organic contaminants correlate well with their P_L° values on a logarithm scale (Zhang *et al.*, 2009; Tseng *et al.*, 2014; Wu *et al.*, 2014b). Although PAH isomers are emitted primarily in gaseous phase during high-temperature pyrolysis processes, gaseous PAHs are quickly partitioned into particle matter by condensation and adsorption, and the equilibrium between gas and particle phases is reached within a short time (Liang and Pankow, 1996; Spezzano *et al.*, 2009). PC 2 loading of the rotated component matrix was potentially distinguished as cooking- and traffic emission-related PAHs. Briefly, mass concentrations of BghiP, BbF, IcdP, BeP, BaA, and two alkylated PAHs negatively correlated with their PC 2 loading values; however, the concentrations of Ant, BaP, DahA, BkF, Flu, Chr, and Phe positively correlated to their PC 2 loadings (Fig. 3(c)). As discussed above, BghiP, IcdP and BeP would be expected from cooking activities (See *et al.*, 2006), and Flu and Pyr would be considered the markers of vehicle-associated mobile sources. Therefore, PC 2 represented the emission sources of PAHs.

Occupational Inhalation Exposure

The IEL of the night market staff to the BaP-adjusted PAHs was $1.75 \pm 0.878 \mu\text{g day}^{-1}$, with a range of 0.451–3.43 $\mu\text{g day}^{-1}$, indicating high potential exposure risk due to the occupation. The highest IEL value was investigated

at Datong Square, and the lowest value of IEL was found at the northern gate of the western campus of the University of Science and Technology of China, where the highest and the lowest concentrations of Σ PAH were obtained, respectively. The daily inhalation dose in the present study is comparable to that for the population in the night market in Taiwan, which was significantly affected by grilling practices (Kuo *et al.*, 2005). A previous study also showed temple workers in Taiwan with a comparable daily inhalation dose (0.076 $\mu\text{g kg day}^{-1}$ at the 50th percentile) (Chiang *et al.*, 2009). Notably, only inhalation exposure was considered in the present study; ingestion of and dermal contact with particle-bound PAHs would potentially be expected to highly contribute to total human exposure to airborne PAHs (Chiang *et al.*, 2009; Chen *et al.*, 2012).

In the present study, we estimated the potential ILCR by the occupational exposure to PAHs of 1, 3, 5, 10 and 20 years (Fig. 4). Our results suggest that the occupational inhalation risk of the workers with 1 year of service time was primarily less than the acceptable risk level of 10^{-6} (1.3×10^{-7} – 8.4×10^{-7}) for all sampling sites, while the risk was greater than 10^{-6} in five sites for the people who had worked for 3 years (Fig. 4(a)). However, all ILCR values were higher than 10^{-6} but lower than the priority risk level (10^{-4}) for the staff with 20 years of work experience (2.5×10^{-6} – 1.7×10^{-5}). However, the people at control site would be expected to have a low inhalation exposure risk because the ILCR values were less than 10^{-6} , except for 20-year inhalation exposure, which was 1.3×10^{-6} . These results indicate the potential carcinogenic risk for the workers in the night markets of Hefei City, a typical metropolis in China.

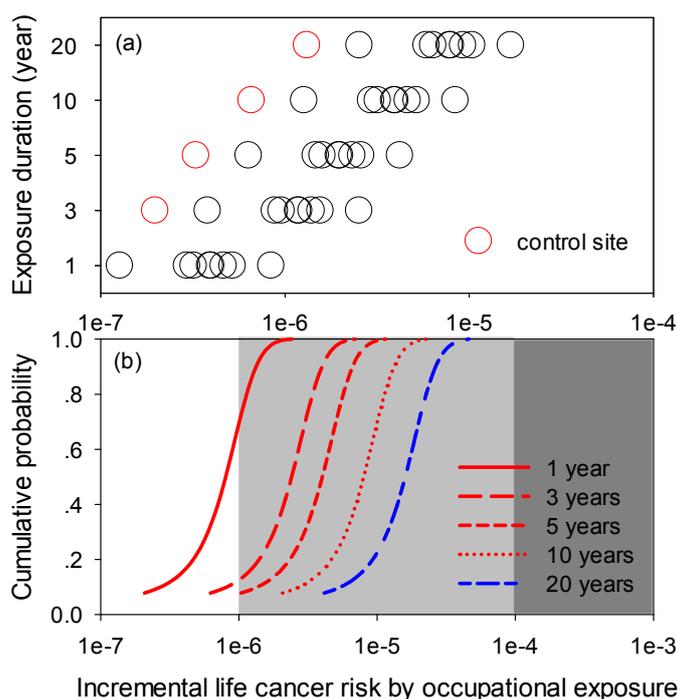


Fig. 4. The incremental life cancer risk (ILCR) (a) and cumulative probability of ILCR (b) of 1, 3, 5, 10, 20 years-occupational exposure to PAHs for workers in the night markets.

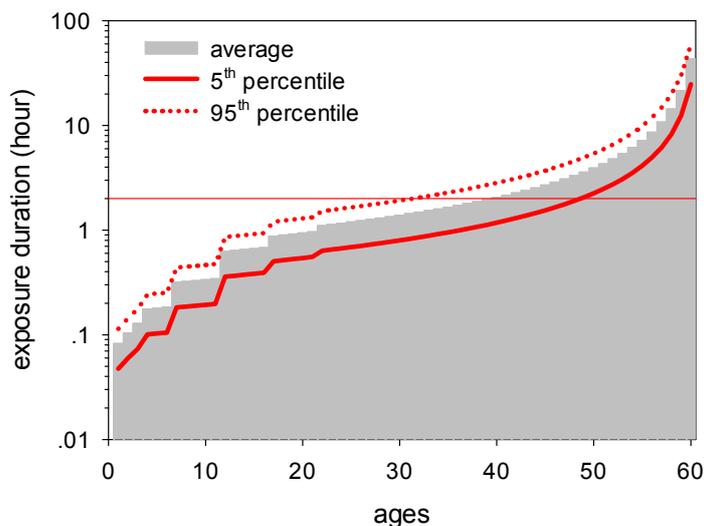


Fig. 5. Predicted maximum consumption time for consumers of different ages.

The highest and lowest ILCR values were found at Datong Square and at the northern gate of the western campus of the University of Science and Technology of China, where the highest and lowest concentrations of Σ PAH were obtained, respectively. The probability of worker exposure to PM_{10} -bound PAHs in night markets was also evaluated by combining the probit mode with the estimated ILCR values, indicating that these people will be exposed to acceptable risk levels of airborne PAHs, with probabilities of 0.26% for 1 year, 66.0% for 3 years, 85.3% for 5 years, 93.7% for 10 years and 96.1% for 20 years (Fig. 4(b)).

Combining the acceptable risk level, t_{max} was estimated

for the customers with ages from 1 to 55 years old (Fig. 5). The acceptable t_{max} was 2.5–7.1 minutes for one-year-old infants, 3.9–11 minutes for 3-year-old toddlers, 5.5–29 minutes for 5–10-year-old children, 20–57 minutes for 15-year-old adolescents, 0.48–1.34 hours for 20-year-olds, and 1.99–5.60 hours for 50-year-old adults. Therefore, the estimated consumption time under the acceptable risk level increased with an increase in age, and infants, toddlers and children are vulnerable to a high exposure risk by inhalation of PM_{10} -bound PAHs. Specifically, the maximum exposure time in the night markets for the customers under 25 years old should be less than 1 hour.

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